

Microphotoluminescence study of disorder in ferromagnetic (Cd,Mn)Te quantum well.

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Microphotoluminescence mapping experiments were performed on a modulation doped (Cd,Mn)Te quantum well exhibiting carrier induced ferromagnetism. The zero field splitting that reveals the presence of a spontaneous magnetization in the low-temperature phase, is measured locally; its fluctuations are compared to those of the spin content and of the carrier density, also measured spectroscopically in the same run. We show that the fluctuations of the carrier density are the main mechanism responsible for the fluctuations of the spontaneous magnetization in the ferromagnetic phase, while those of the Mn spin density have no detectable effect at this scale of observation.

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The realization of carrier-induced ferromagnetism [1, 2], with possible applications in such rapidly developing areas as "spintronics", has boosted the interest in the study of diluted magnetic semiconductors (DMS) with a large density of carriers. The experimental work has been mainly focused on (Ga,Mn)As, resulting in a rapid advancement in the control of defects in layers grown by molecular beam epitaxy (MBE), and the achievement of relatively high values of the critical temperature T_c . Some devices have already been implemented [3]. From the theoretical point of view, a very simple mean field model (Zener model) appears to be surprisingly successful [1, 4]. One main advantage of this model is that it uses parameters, which are deduced from spectroscopic studies. However, it neglects disorder and fluctuations.

Disorder in these materials has many sources. The solubility of Mn in GaAs is low, so that layers have to be grown at low temperature, resulting in a large amount of structural defects; some of them (As antisites, Mn interstitials) can be eliminated at least partially by a careful adjustment of the III/V ratio and post-growth annealing, what leads to higher values of the conductivity and of T_c [5, 6, 7]. However, disorder is intrinsically present in doped DMS, and it is widely recognized that this is a crucial issue for understanding carrier-induced ferromagnetism in semiconductors. Thermal fluctuations have been early incorporated in the mean field model by considering elementary excitations [8]. The random distribution of the localized spins results in spin-glass like behaviours in undoped DMS [9], an effect which should be even enhanced by the oscillatory character of the RKKY interaction mediated by free carriers. Electrical doping of semiconductors also involves a random distribution of electrically active impurities. The Zener model assumes the presence of free carriers, and the opposite limit of strongly localized carriers (where magnetic polarons form) has been considered [10, 11]. However,

actual samples are on both sides of the metal-insulator transition, and close to it. More recent models aim at fully incorporating disorder [12, 13, 14, 15]. They will help to understand, what must be considered as a specific consequence of disorder, and where its effects should be searched experimentally.

Experimental studies of disorder in doped DMS's are scarce. In addition, several features of carrier induced ferromagnetism in DMS's, which have been sometimes considered as due to disorder, are already present in the Zener model. As a first example, if the two subsystems (localized spins and free carriers) are treated separately, one of them will saturate first; in the case of DMS's, the carrier density is small and the carrier gas is fully polarized while the magnetic ions spin system is far from saturation; as a result, the magnetization follows a Brillouin function, which increases when a magnetic field is applied at constant temperature [16], or when the temperature is decreased at constant (even zero) field; in the latter case, an upward curvature is expected [17] which in this case has nothing to do with disorder. Another example is the threshold in the dependence of T_c on carrier density or spin density, which is reproduced [2, 17, 18] if the short-range spin-spin interactions are introduced in the mean field model (in a phenomenological way, but without adjustable parameter).

A direct effect of disorder is the reduced value of the spontaneous magnetization. This is a very common observation in ferromagnetic DMS's, which has been rapidly attributed to the presence of uncorrelated spins [1]. In (Ga,Mn)As, such spins are expected in those parts of the material with smaller Mn content, and hence smaller spin and carrier density. The effect is particularly strong in samples, which are on the insulating side of the metal insulator transition. The study of nitrogen-doped (Zn,Mn)Te [18] gives further insight into the mechanism, since in that case Mn is an isoelectronic impurity

and carriers originate from the nitrogen acceptor. It was clearly observed that the measured spontaneous magnetization closely matches the prediction from the mean field model in metallic samples, while it is significantly smaller in the insulating ones [18]. This suggests that electrostatic disorder plays the most significant role (It should be kept in mind however that the presence of the Mn makes the Mott critical density to increase: this effect is suppressed by applying a magnetic field, which confirms the role of spin fluctuations [18]). Another confirmation comes from the study of modulation-doped (Cd,Mn)Te quantum wells (QW). The measured magnetization was found [19, 20] to be proportional to both the spin density x_{eff} and the carrier density p , in agreement with the mean field model - in spite of the low values of p . This observation can be ascribed to the strong reduction of electrostatic disorder, achieved by using remote doping in such structures, as compared to (Zn,Mn)Te layers.

As the magnetization in a (Cd,Mn)Te QW is usually deduced from the giant Zeeman splitting of the semiconductor's photoluminescence (PL), local measurements can be performed using micro-PL. We present here a study of the static fluctuations of the local, spontaneous magnetization in p-doped (Cd,Mn)Te QWs, i.e., in a DMS structure where (i) thermal fluctuations are minimized (the highest critical temperature observed for such systems was 6.7K), (ii) electrostatic disorder is smaller than in thick layers (thanks to remote doping), and (iii) magnetic disorder is due to the random distribution of Mn atoms. We examine how these fluctuations correlate to those of the spin density and of the carrier density, also deduced from micro-PL spectra from the same run.

Micro-PL was measured in a specially designed set-up with a microscope lens immersed in the pumped helium bath of a superconducting magnet [21], at temperatures from 4.2 K down to 1.5 K. PL maps of a 32×32 micrometer wide square area were recorded by placing a 10mm thick plane-parallel quartz plate between the sample and the first lens of the optical system and rotating the plate around two perpendicular axes. PL was excited using a semiconductor diode laser (680 nm) focused to a spot of diameter smaller than 0.5 micrometer, and recorded using a CCD camera at the exit plane of a spectrograph.

The sample was a single, 8 nm wide, (Cd,Mn)Te QW embedded between (Cd,Mg)Te barriers (27% Mg), grown by MBE technique on a (001) oriented, 4% (Cd,Zn)Te substrate. The QW contained 4.65% of Mn, as determined from reflectivity spectra in magnetic fields up to 4.5 T, by fitting the Zeeman splitting with a modified Brillouin function [22] (not shown) with the parameter $x_{eff} = 2.74\%$. The QW was covered by a 25 nm thick cap layer, thin enough so that a hole gas of density about $3 \times 10^{11} \text{cm}^{-2}$ was generated by acceptor surface states [24]. The use of surface states as a source of holes - instead of nitrogen doping - allows us to grow the sample at the optimal temperature of 280°C, so that pseudo-

smooth interfaces are realized [25]. The white light illumination in such experiments efficiently depletes the QW from its hole gas [23].

The general properties of PL from a p-doped (Cd,Mn)Te QW have been described in detail in ref. 26. We recall here the minimum information needed to extract the local values of p and x_{eff} and the spontaneous magnetization M_0 from micro-PL data, as summarized schematically in the central part of Fig.1.

At small spin splitting (either spontaneous or field induced), PL is due to the recombination of the positively charged exciton, X^+ . It takes place in σ^+ polarization if the recombined hole belongs to the majority spin sub-band, and in σ^- for holes with minority spin (all being defined with respect to the local magnetization). The splitting between the two lines is the so-called giant Zeeman splitting; it is proportional to the local magnetization. In a QW with high enough values of p and x_{eff} , the onset of ferromagnetic ordering is witnessed by a zero-field Zeeman splitting of the PL line, Fig. 1a [2, 17], which signs the presence of a spontaneous magnetization inside the QW, at temperatures below T_c . For the present sample, we determined $T_c = 2.5$ K. In the paramagnetic phase, one observes a critical divergence of the field-induced PL splitting when decreasing the temperature, which follows a critical law, $\chi(T) = C/(T - T_{CW})$. No such effects are detected in undoped structures or under white-light illumination (which significantly reduces the hole density in the QW). In samples with relatively low Mn content, such as here, the two characteristic temperatures T_c and T_{CW} coincide. Note that, due to the anisotropy of the heavy hole states, the exchange field is perpendicular to the QW plane. Thus it corresponds to the normal Faraday configuration and circular polarizations of emitted light. However, if the signal is averaged over areas of magnetic domains of both possible orientations, the observed PL is not circularly polarized [20]. It was shown [27] that even in the micro-PL experiment with resolution better than 1 μm , PL is averaged over different domains. The zero-field splitting measured in micro-PL gives us the average value of the local magnetization within the domains present in the imaged spot.

When the spin splitting exceeds the X^+ binding energy, a double line is observed. The high energy component has the ground state of the hole gas as a final state, and it shows a clear phonon replica. The low-energy component leaves the hole gas in an excited state: the energy of this final state monotonously increases with p (from about 2.5 meV for vanishing values of p , to the Fermi energy for high enough values of p [26]), so that the observed splitting is a good measure of the local carrier density (which was calibrated in Ref. 26 for carrier densities in same the range as in the present study). This is illustrated in Fig.1b, for two different values of the carrier density (as determined from the well known Moss- Burstein shift, i.e., the shift between PL and ab-

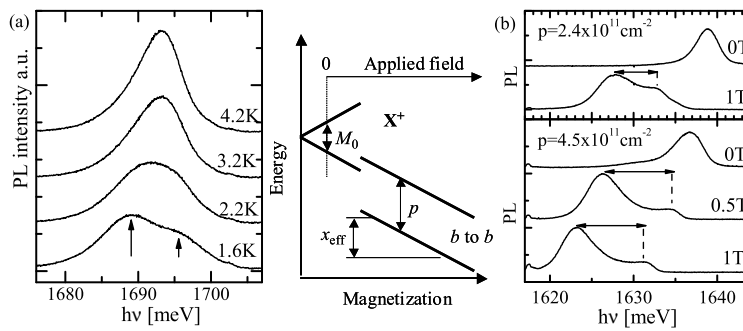


FIG. 1: Characteristic features of PL from a p-doped (Cd,Mn)Te QW. (a) PL at zero field, from the sample with 4.65% Mn, at different temperatures. The arrows show the zero field splitting, due to the giant Zeeman splitting of the charged exciton (X^+ in the central scheme) induced by the spontaneous magnetization M_0 ; (b) PL from a QW with 1.4% Mn, at two values of the carrier density (as deduced from the Moss-Burstein shift). The double arrows on the double line (which is close to a band to band transition, b to b in the central scheme) indicate the splitting used to measure the carrier density.

sorption, which was calibrated in [26]) in a sample with 1.4% Mn (so that absorption could be measured through the $\text{Cd}_{0.96}\text{Zn}_{0.04}\text{Te}$ substrate). This splitting does not depend on temperature (as soon as the spin splitting is large enough), which makes it easy to distinguish from the spontaneous Zeeman effect. Destabilization of the X^+ by the giant Zeeman effect due to spontaneous magnetization was observed, e.g., at low enough temperature. In the present study, the spontaneous magnetization was always smaller than the value needed to destabilize the X^+ .

Finally, the local value of x_{eff} was deduced from the field-induced shift between two values of the applied field, 1 and 3 T. Note that for such values of the spin splitting, there is no doubt that the hole gas is fully polarized, so that carrier-carrier interactions do not change with field and temperature. At incomplete spin polarisation, carrier-carrier interactions leads to additional shift of the PL line [23, 26]. Also, the effect of the exchange field is negligibly small at such field values.

Figure 2 shows typical maps of the different parameters describing the PL spectra, obtained with sub-micrometer spatial resolution, and selected from the ensemble of measurements performed on different areas of the sample. PL was excited directly in the QW (at 680 nm), and the excitation power was checked to be low enough to avoid significant heating of the Mn system. Fig. 2a shows the zero-field PL splitting at 1.8 K, which is the measure of the local magnetization within the domains. Fig. 2b shows the splitting between the two components of the σ^+ PL line at $B = 1$ T, which depends on the local hole density. Other maps (not shown here) were plotted for the Zeeman shift between 1 T and 3 T, which corresponds to the spin density, and for the total PL intensity.

All maps (except the map of total intensity) exhibit significant, irregular, spatial fluctuations. Their characteristic scale is a few micrometers. These fluctuations of the spectroscopic parameters reveal fluctuations of the

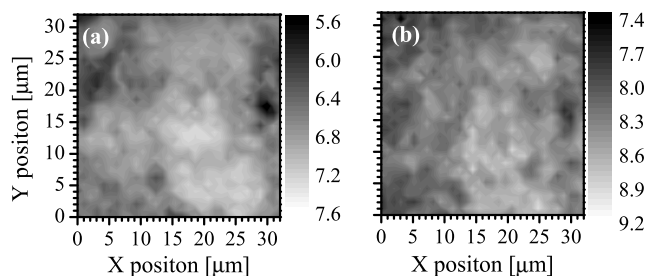


FIG. 2: Micro-PL maps of (a) zero field splitting (in meV) and (b) splitting between two components of a σ^+ PL line at $B = 1$ T (in meV)

local magnetization, hole density and Mn content, respectively, for the maps of zero-field splitting, splitting at $B = 1$ T, and high-field Zeeman shift.

In order to decide what is the nature and the origin of the fluctuations of the spontaneous magnetization, we analyzed the correlations with the fluctuations of the Mn content (high field Zeeman shift, fig 3a) and hole density (high-field splitting, fig 3b). We have also checked (not shown) that there is no correlation between the fluctuations of carrier density and those of spin density. Only figure 3b evidences a meaningful correlation. On the 1024 points of Fig. 3b, the Pearson's correlation coefficient, defined as $R = \langle xy \rangle / \sqrt{\langle x^2 \rangle \langle y^2 \rangle}$ where $\langle x^2 \rangle$ is the centered second moment of x , is $\approx 58\%$; for such statistics, a correlation coefficient larger than 8% ensures a significance level of 0.01 (i.e., 99% confidence). This demonstrates that the main mechanism responsible for the fluctuations of spontaneous magnetization is the presence of fluctuations of the hole density. On the other hand, no clear correlation appears in Fig. 3a (on 256 points, $R \approx 7\%$, much less than the threshold of 16%). Note that this may simply indicate that the distribution of spin density is too narrow to have any detectable influence on the spontaneous magnetization. On a macroscopic scale, the

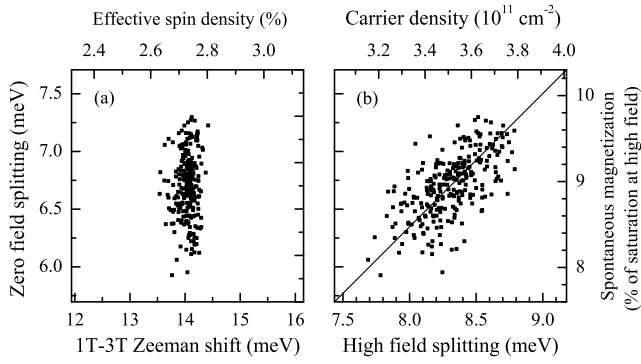


FIG. 3: Plot of the zero field splitting as a function of (a) the Zeeman shift between 1 and 3 T, and (b) the splitting measured at high field, using data from Fig. 2. The three scales have been chosen so as to show a $\pm 15\%$ variation around the mean value. The top scales are deduced from the measured data using (a) available parameters of the giant Zeeman effect in CdMnTe [22] and (b) the calibration of a hole gas in a CdMnTe QW [26]. The magnetization scale (right scale) uses the giant Zeeman effect measured at large field on a large area of the present sample. The solid line shows a linear dependence of the spontaneous magnetization on carrier density.

spontaneous magnetization was found to be proportional to both the effective spin density and the carrier density [19,20]. This dependence is shown in Fig. 3b by a solid line. It agrees with the correlation existing between the local values of the two parameters.

Such fluctuations of the carrier density might result from electrostatic fluctuations, or fluctuations of the QW parameters such as strain and QW width. Fluctuations of the QW width should have rather similar effects on the carriers and on the excitons. They are probably small in the case of samples grown at 280°C , where pseudo-smooth interfaces are found (i.e., the characteristic scale is smaller than the exciton coherence length) [25]. Strain fluctuations are also minimized by a careful design of the sample structure, so that the whole layer is coherently strained to the substrate. The main source of fluctuations of carrier density is thus probably the fluctuations of electrostatic potential, due to the distribution of ionized acceptors. One generally considers [28] that the typical spatial scale of these fluctuations is determined by two parameters: the distance from the QW to the doping layer (in our case, with doping from surface states, this is the thickness of the cap layer, 25 nm), and the inverse of the Fermi wavevector (about 12 nm for $p = 3 \times 10^{11}\text{cm}^{-2}$).

To conclude, in spite of a rather low spatial resolution ($0.5\ \mu\text{m}$) inherent to a micro-PL set-up, the fluctuations of the carrier density in a modulation-doped p-type $\text{Cd}_{0.96}\text{Mn}_{0.04}\text{Te}$ QW have been imaged, and their role on the spontaneous magnetization due to carrier induced magnetism demonstrated. The effect of spin density fluctuations

is too small to be detected. We may anticipate that the role of electrostatic fluctuations should be even more dominant in a thick layer, where the acceptors are present in the DMS layer. Such an optical determination of fluctuations should be feasible also in $(\text{Cd,Mn})\text{Te}$ QWs with a higher Mn content, where stronger disorder seems to induce a difference between the Curie-Weiss temperature measured in the paramagnetic phase under applied field, and the critical temperature at zero field [29]

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